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A precision determination of the half-life of Radium C′

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With 5 figures in the text

Abstract: The half-life of Radium C′ has been determined using a 20-channel time analyzer to measure the time distribution of α-pulses from a proportional counter, which contains a small radon capillary, relative to a preceding β-pulse from a set of G–M-counters surrounding the α-counter. The value obtained for the half-life is 163.7 ± 0.2 μsec, the standard error being that due to the statistical fluctuations of the experimental points, increased by a factor of $\sqrt{2}$ in order to take account of possible unknown sources of accidental or systematic error. The value is not inconsistent with previous values obtained with the method of delayed coincidences, but seems outside the experimental errors of values obtained with the method of variable resolution. It is suggested that systematic errors may be present in values obtained by this latter method.

A. Introduction

Radium C′, a member of the Ra-series, is known to decay with a very short half-life to Radium D by emission of a 7.68 MeV α-particle. It is formed by the β-decay of Radium C, which has a half-life of 19.7 min.

RaC′ was first estimated from the Geiger Nuttall law to have a half-life of about 10^{-6} sec. Indirect experiments by recoil methods seemed to confirm a value of this order of magnitude. The first direct measurements were accomplished by JACOBSEN (1) who observed the time difference between α-ray and β-ray pulses with a double coil oscilloscope. He found the half-life to be of the order of 10^{-4} sec, in good agreement with the Gamow formula for α-decay. Later the half-life of RaC′ has been measured by the coincidence method of variable resolution and by the method of delayed coincidences. The first method was used almost simultaneously by DUNWORTH (2) and ROTBLAT (3) yielding values of 150 ± 20 μsec. and 140 ± 15 μsec. respectively. The second author later improved his accuracy (4) and obtained 145 ± 5 μsec. Later measurements by the same method by WARD (5), who used a single G–M counter to detect both the α- and β-particles, and of BUNYAN, LUNDBY, WARD and WALKER (6), yielded values in good agreement with ROTBLAT's later value. The method of delayed coincidences has been used by JACOBSEN and SIGURGEIRSSON (7), who obtained a half-life of 155 ± 5 μsec.

A multichannel differential method has been used by BERTHELOT, CHAMINADE and WAHL at the laboratory of CEA in Paris (8). The principles of their measurement are similar to those of the present experiment. Their result is in good agreement with the value of JACOBSEN and SIGURGEIRSSON and is of comparable accuracy.

Due to its general availability and the relative ease of the experiment RaC′ has been used extensively as a standard substance to check the proper functioning of equipment and the validity of methods intended for the determination of the half-lives of other short-lived radioactive substances. A systematic error in the accepted value for the half-life of RaC′ may therefore reproduce itself in the half-lives of other substances, which have been measured by the same methods. It was therefore considered to be of some value to make a precision determination of the half-life of RaC′.

B. Apparatus

General

The important part of the apparatus used in the present experiment is a 20-channel time-analyzer originally developed for use with a neutron time of flight spectrometer. The apparatus is similar in principle to the time analyzers built for neutron time of flight experiments by BACHER, BAKER, MCDANIEL and JONES (9, 10) and by RAINWATER et al. (11, 12). A similar apparatus was used by CASSELS and LATHAM (13) to determine the half-life of 0.87 sec ^{6}He, activated in the cyclotron. Another time-analyzer intended for determination of very short half-lives, constructed by CHAMINADE at the laboratory of CEA, has already been mentioned (8). Only a general description and a block diagram, Fig. 1, will be given of the instrument in connection with the present investigation. The essential part of the instrument is an electronic switch with 20 positions which connects the input circuit for the pulses, whose time distribution is to be studied, in turn to 20 channels. Each channel contains a scale of 4 and a mechanical register. The switch is operated at equal intervals by pulses from an oscillator. When used to determine the half-life of a substance in a radioactive decay series, such as RaC′, the switch is normally in the zero-position. When a triggering pulse arrives, indicating the formation of an atom of the substance, the switch is operated in steps by equidistant pulses until it has returned to the zero-position after one complete revolution.

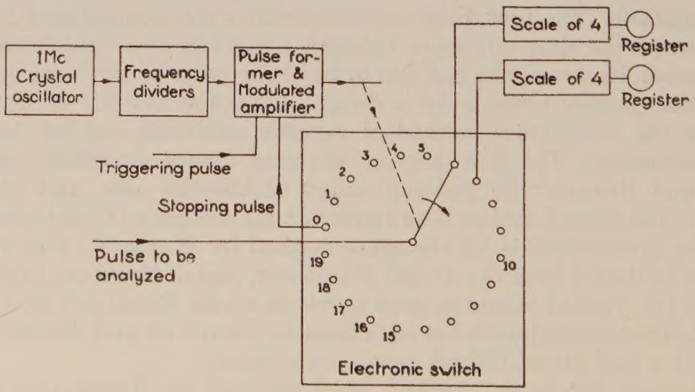

Fig. 1. Block diagram of 20-channel time analyzer.

Definition of the channels

The essential part of the electronic switch is a system which generates rectangular gating pulses. These open the input of the scale of 4 in one channel at a time to the pulses which are to be analyzed. The rectangular pulses will in practice always have finite rise and decay time, the effective width of each channel being defined by the forms of the front and rear edges of the gating pulse and by the sensitivity level of the first scale of 2 in the channel. These factors depend on tube characteristics and stray capacities and will introduce a considerable inaccuracy, if the rise time of the pulses is not very small compared to the width of the channels. In the present system the rise time of the gating pulses is about 2 μsec, and difficulties would probably be encountered if precision measurements of the 150 μsec half-life of RaC′ were tried without improving the system. One obvious improvement would be to decrease the rise time of the gating pulses by using small load resistors and high currents.

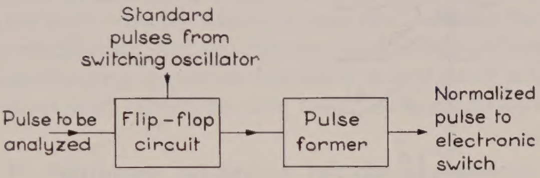
Standard pulses from switching oscillator
Pulse to be analyzed
Flip-flop circuit
Pulse former
Normalized pulse to electronic switch

Fig. 2. Block diagram of prearranging circuit.

However other difficulties would probably arise in this case due to the decreased tube life and increased heating of the components. Large modifications of the entire system would be necessary. A more satisfactory solution in our case was found in the use of the prearranging circuit, shown in block diagram Fig. 2. The pulses to be analyzed are not fed to the electronic switch directly but to a flip-flop circuit. Standard pulses from the oscillator which operates the electronic switch are fed to the other input of the flip-flop circuit, which will be "flopped" back by the first standard pulse after the input pulse. The rear edge of the square pulse generated by the circuit is differentiated, amplified and fed to the electronic switch. The function of the circuit is thus to replace the original pulse, which may sometimes coincide with the edges of the gating pulses, by a pulse which has a fixed position with respect to the gating pulses, so timed that it arrives after the switching action has been completed. This procedure completely eliminates the inaccuracy of the definition of the channels due to tube characteristics and other nonconstant factors. The widths will be determined entirely by the separation between the standard pulses from the switching oscillator. During the main part of the measurements the pulses, which trigger the electronic switch, were supplied by a crystal-controlled, 1000 kc oscillator with associated frequency dividers and pulse-forming circuits. The interval between pulses can be chosen between 2 and 200 μsec. The pulses are blocked from the switch by a valve, which is normally cut off. A triggering pulse opens the valve, and the switch is turned in steps until it has returned to the zero position, when the valve is cut off and the switch stops.

Another system which was used for some measurements uses a blocking oscillator for supplying the switching pulses. The oscillator is normally held inoperative. When a triggering pulse arrives the oscillator starts turning the switch, until it has returned to zero, when the oscillator is stopped. This system was less suited for precision measurements because of the frequency drift and transient behaviour of the blocking oscillator.

C. Resolution

When a time distribution f(t) is measured it will always be modified by the limited resolution of the apparatus by which it is analyzed. The effect of this resolution is the smearing of the original time distribution by a resolution function $K(t, \tau)$, so that the measured time distribution is of the form: $F(t) = \int f(\tau) \, . \, K(t, \tau) d\tau$. The form of the resolution function depends on the method used for the analysis. Some important forms of the resolution function are shown in Fig. 3. The coincidence method of variable resolution (Fig. 3 A) has a resolution function of rectangular form. As the resolving time T is changed, the time integral of the original distribution will be plotted. The method is often called the integration method. The method of delayed coincidences (3 B) measures the average value of the time distribution over a small

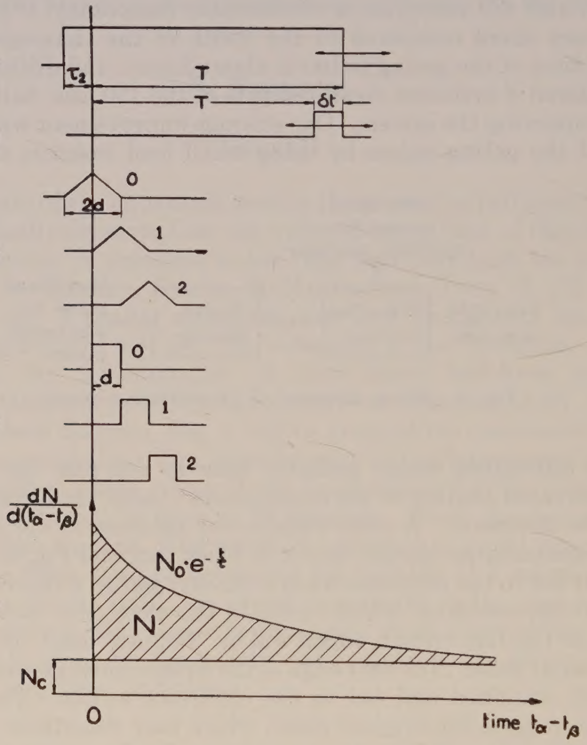
3A
τ_2
T
T
δt
3B
0
2d
3C
1
2
0
d
3D
1
2
$\frac{dN}{d(t_\alpha - t_\beta)}$
$N_0 \cdot e^{-\frac{t}{\tau}}$
3E
N
N_c
0
time $t_\alpha - t_\beta$

Fig. 3 A—D. Resolution functions of various circuits used for measuring short half-lives. A. Coincidence method of variable resolution. B. Delayed coincidence method. C. Multichannel differential analyzer with *gated* oscillator. D. Multichannel differential analyzer with *triggered* oscillator.

Fig. 3 E. Distribution of delays between α- and β-particles from a radon sample. N = total number of particles in the exponential component. N_c = chance coincidences.

interval δt. As the delay T is changed, the time distribution will be plotted point by point. The method is often called the differential method. The present method is a multichannel differential method where a number of points of the time distribution are simultaneously measured in different channels, which have resolution functions of the general form 3 A but with different delays, T. The exact form of the resolution function depends on the principle of the apparatus used. During the main part of the present experiment a continuously running oscillator with pulse distance d was used to switch the electronic switch. In this case the phases of both the triggering and analyzed pulses with respect to the switching pulses are quite arbitrary. The effect is a resolution function of triangular form of unit height and base $2 . d$ (Fig. 3 C). Adjoining channels overlap by the amount d. On the contrary the blocking oscillator used during part of the experiment is locked in phase to the triggering pulse and only the phase of the analyzed pulse is arbitrary. In this case the resolution function will be of rectangular form of unit height and of width d (Fig. 3 D). With a multi-channel system in general the resolution functions of the different channels cannot be relied upon to be strictly identical and the channels have to be calibrated with a known time distribution, e.g. the constant distribution of random coincidences. The effect of the prearranging circuit as described in section B is to make the resolution functions identical and eliminates the need for a calibration of the channels.

D. Optimum conditions for the experiment

In order to determine the half-life of RaC′, one studies the time distribution of the α-pulses which indicate the decay of a RaC′-atom, relative to the β-pulses, which indicate the birth of the atom. As the decay is exponential, the time distribution should show an exponential behaviour for positive time-differences, superimposed upon a constant background of random α-pulses which are not related to the particular β-pulse taken as zero-time (Fig. 3 E). In an efficient experiment the rate of counting in the exponential part of the distribution N should be large, and the ratio $1 : \lambda$ of background N_c to the initial value N_0 of the exponential part of the distribution should be small.

Part of the background counts are due to the natural background of the counters. With the high counting rate used in the present investigation this contribution to the random coincidences may be neglected. If — as in the present measurements — radon is used as the source for RaC′, the α-pulses from RaA and radon itself and the β-pulses from RaB will add to the random coincidence rate. The fact that these radiations are less penetrating than the rays from RaC and RaC′, makes the use of appropriate filters advantageous.

The counting rate in the exponential part of the time distribution is proportional to the source strength. Except for the small part caused by the natural background of the counters the background rate is proportional to the square of the source strength, and the ratio $1 : \lambda$ is proportional to the source strength. Peierls (14) has calculated the accuracy that can be obtained in the determination of the half-life from a time distribution of the form shown in Fig. 3 E. If the only sources of error are the statistical fluctuations of the number of counts, the relative standard error is $1/\sqrt{N}$ for no background, $(1 : \lambda = 0)$. The standard error increases with $1 : \lambda$. If the source strength is increased the gain in accuracy due to the higher counting rate outweighs the loss due to the higher relative background. For best accuracy a rather strong source should therefore be used.

E. Measurements

The experimental arrangement is shown in principle in fig. 4. The α-counter used was a thin-walled counter of the same type as a β-counter of conventional design, but filled to about 1 atmosphere of pure argon and operated at 1000–1200 volts as a proportional counter for α-rays. The counter was mounted directly on the input of a linear amplifier having a gain of 10^4. The amplified pulses are discriminated, and the output of the discriminator operates a scale of 64 and the time analyzer.

The α-counter is surrounded by a battery of 6 thin-walled β-counters operated in parallel. A β-ray from the source passes through the thin walls of the α-counter and fires one of the surrounding β-counters. The β-pulses pass through a preamplifier to a discriminator, the output of which operates a scale of 64 and the time analyzer.

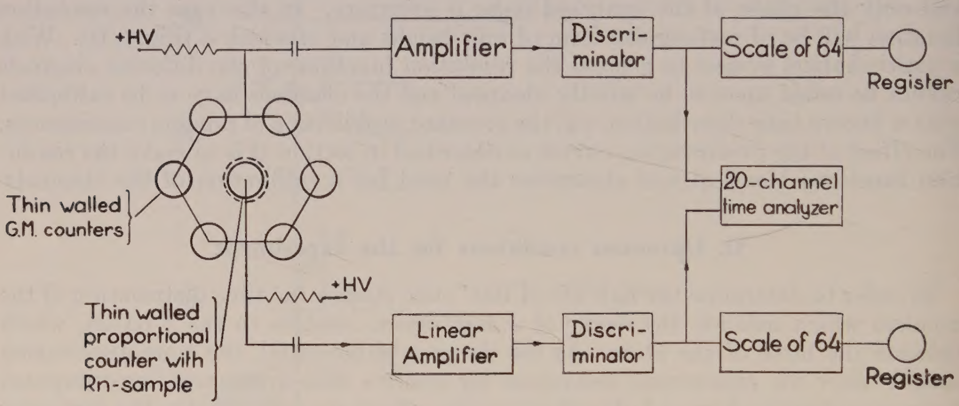

Fig. 4. Block diagram of the experimental setup for determining the half-life of RaC′.

The whole assembly is surrounded by lead bricks in order to decrease the natural background of the counters.

The β-pulses trigger the oscillator of the time analyzer and start the electronic switch. The α-pulses are fed to the prearranging circuit and are analyzed by the electronic switch.

The channel widths used in the present experiment were chosen from two considerations. In the first place the channel widths should not be larger than about 0.5 half-lives, as otherwise the corrections for the finite distance between experimental points would be too large with the method of treatment of the data. Also the accuracy would be decreased, as details of the time distribution would be lost. On the other hand the exponential decay should be measured over a sufficient number of half-lives, so that the background of chance coincidences could be determined by extrapolation with good accuracy. In order to avoid systematic errors the channel-widths were varied within the limits set by these two requirements during the measurements.

The final measurements were made in 3 series of runs. The length of each series was limited by the decay of the radon source (3.8 days half-life). Each series was subdivided into a number of runs. After each run the mechanical registers of the time analyzer were read and the proper functioning of the apparatus was tested with artificial pulses.

Series A consisted of 6 runs between 19 and 23 hours measuring time, taken during the time December 23, 1948 to January 3, 1949. The total efficient measuring time was 123 hours. The radon gas used for the source was introduced into the α-counter, so that an active layer, which contained RaC, was deposited on the walls. With this arrangement there is no filtering of the α-rays. The time analyzer was in a preliminary state of construction and contained only 10 channels. The blocking oscillator mentioned in the last paragraph of section B was used to trigger the electronic switch. The channel width was 50 μsec.

Series B consisted of 19 runs of between $1\,{}^1/_2$ and 51 hours length, taken in the time November 23 to December 4, 1949. 2 runs had however to be rejected because of an error in the setting of the frequency. The total efficient measuring time was about 200 hours. For this series, the radon source was contained in a glass capillary of a thickness, such that the emerging α-rays from RaC′ had a residual range of about 5 mm in air. All α-particles from Rn and RaA were stopped, but the number of emerging RaC′ α-rays was also considerably reduced, as most RaC′ α-particles which were emitted at an oblique angle with the glass were also absorbed. The counting rate for α-particles was therefore only about a third of what would have been the case if a more suitable wall thickness had been chosen. — The channel width used during series B was 77 μsec.

Series C consisted of 14 usable runs of between 2 and 102 hours in length measured between December 10 and 29, 1949. The subdivision in runs is shown in detail in Table 2. The effective measuring time was 350 hours. Series C contributed the bulk of the experimental data for the determination of the half-life. The same sort of radon source capillary was used as in series B, but the wall thickness of the capillary was somewhat less, leaving a residual range of about 20 mm for the α-particles from RaC′. The thickness was still sufficient to stop effectively the α-particles from Rn and RaA, and the loss of RaC′ α-particles penetrating the glass at an oblique angle was much less than in series B. The initial strength of the source was greater than in the previous series of measurements and the decay of the source was followed for a longer time, so that better statistics were collected.

The initial strength of the source was estimated from the initial counting rate for α-particles, 230 pulses per sec., to be about 10^{-8} curies. There would still be something to be gained by increasing the source strength above this value, as the increase in counting rate would outweigh the accompanying decrease in accuracy due to the increased chance coincidence rate. However with the apparatus used for the present experiment the maximum usable counting rate was limited by the resolving power of the mechanical registers (see below, Section G).

The channel widths used during series C are shown in Table 2, column IV.

For the measurements of series B and C the crystal controlled oscillator and blocking circuit described in Section B were used to trigger the electronic switch.

F. Treatment of experimental data

During the measurements no attempt was made to measure absolute counting rates or to keep the efficiency of the counters constant from run to run. The treatment of the experimental data is therefore limited to the determination of factors which do not depend upon absolute counting rates, but only on the ratio of counting rates in different channels. The most important of these factors is the half-life of the

Table 1

Run No. 4, Series C. (12/12 19.09 — 13/12 11.30) = 16^h21^m

Channel width: $d = 77$ μsec.
Total number of α-counts: 193.578 × 64
Total number of β-counts: 100.092 × 64

Ch. No. I	Exp. values $\frac{N_i}{4}$ II	Extrap. backgr. $\frac{N_\infty}{4}$ III	Corr. values $\frac{N_i'}{4}$ IV	$\log\frac{N_i'}{4}$ V	$\log\left(\frac{N_i'}{4}, \text{theor. } 164\ \mu\text{s}\right)$ VI	$\log\left(\frac{N_i'}{N_i', \text{theor. } 164\ \mu\text{s}}\right)$ VII	ε VIII
1	106 066	22 170	83 896	4.92376	4.92376	0.00000	0.0009
2	84 304		62 134 [1]	4.79337	4.78242	+0.01095	0.0011
3	65 650		43 480	4.63829	4.64108	−0.00279	0.0015
4	53 566		31 396	4.49693	4.49974	−0.00281	0.0019
5	44 673		22 503	4.35218	4.35840	−0.00622	0.0025
6	38 629		16 459	4.21643	4.21706	−0.00063	0.0032
7	34 042		11 872	4.07445	4.07572	−0.00127	0.0043
8	30 793		8 623	3.93566	3.93438	+0.00128	0.0057
9	28 271		6 101	3.78540	3.79304	−0.00764	0.0080
10	26 619		4 449	3.64826	3.65170	−0.00344	0.010
11	25 414		3 244	3.51108	3.51036	+0.00073	0.014
12	24 595		2 425	3.38471	3.36902	+0.01569	0.019
13	23 933		1 763	3.24625	3.22768	+0.01857	0.026
14	23 414		1 244	3.09482	3.08634	+0.00848	0 038
15	23 078		908	2.95809	2.94500	+0.01309	0.050
16		defective					
17	22 711		541	2.73320	2.66232	+0.07088	0.085
18	22 502		332	2.52114	2.52098	+0.00015	0.14

[1] Interpolated value for channel 2 : 60 450.

Center of gravity: $\frac{s}{d} = \frac{N_1 + 3N_2 + 5N_3 + \ldots. + 19N_{10}}{N_1 + N_2 + N_3 + \ldots. + N_{10}} = 2.69324.$

Correction factor for finite measuring time: 1/0.86521.

Corrected value: $\frac{\tau}{d} = 3.113.$

Correction for finite channel width: −0.050.

Half-life: $(3.113 - 0.050) \cdot 77 \cdot \ln 2 = 163.48$ μsec.

exponential part of the distribution of counts in the different channels, which is identical with the half-life of Radium C′.

The treatment of the data varied somewhat for the 3 series of measurements as the improvements in technique, giving more accurate experimental data, made a more refined treatment necessary. An account will first be given of the procedure used for the data of series C; and a representative run from this series, run No 4, will be used to illustrate the procedure.

The number of counts, N_i registered in the different channels is shown in Table 1, column II. These values were plotted in a diagram against $x = 2^{-(t/T_{1/2})}$. A value of 164 μsec, as indicated by previous measurements, was used for $T_{1/2}$. In this diagram the experimental points should lie on a straight line, which gives a first check on the internal consistency of the data. The intercept of this straight line at $x = 0$ gives the background of chance coincidences N_∞, (column III). The value used for $T_{1/2}$ in the diagram has very little influence on the extrapolation to the background, as the counts in the last channel were always rather close to the background. The experimental values of column II were corrected for the background giving the values. $N_i' = N_i - N_\infty$, of column IV, and the logarithms of these values were found (column V). These should be compared with the linearly decreasing logarithms of column VI, which coincide with column V in channel No 1 and correspond to a true exponential decay with a half-life of 164 μsec. The deviations of the experimental from the theoretical values are found in column VII. Column VIII finally gives the expected standard deviations of the values in column V, calculated by the formula

$$\varepsilon = 0.4343 \frac{\sqrt{N_i + N_\infty}}{N_i - N_\infty}$$

The values of column VII have been plotted in the diagram fig. 5. The lengths of the vertical lines through the experimental points indicate the expected statistical fluctuations according to column VIII. The logarithmic scale on the left, which gives the ordinates in terms of the ratio of experimental to theoretical values has been added for clarity. The diagram of Fig. 5 was used as a second check on the internal consistency of the measurements. The experimental points should lie around a straight line within the statistical fluctuations of the points. A slope of this line indicates a deviation of the true half-live from the assumed value of 164 μsec. A curvature of the experimental curve usually indicated that the background correction should be modified. Departures of individual points from an otherwise smooth curve indicate defects in the channel concerned. An example of this behaviour is given by point No 2 which in this and most other runs shows a systematic departure in the positive direction which seems to be outside the statistical fluctuations. This effect was found to be due to a defective scaler in channel no. 2. For subsequent calculations the values for channel 2 were replaced by values which were interpolated from the plot of Fig. 5. Defects in channels other than no. 2 made the same procedure necessary for about 3 % of the experimental values.

In order to calculate the half-life in an objective and reproducible manner the method of PEIERLS (14) was used. PEIERLS' treatment may be applied to a set of exponentially decaying numbers, taken at equal intervals and corresponding to the same measuring time. These conditions are fulfilled by the values of column IV, after interpolated values have been inserted for the missing or nonusable values from defective channels. PEIERLS' treatment essentially involves the calculation of the center of gravity of the experimental points. A table for the correction factor to be applied for the finite measuring time and a formula for correcting the data for the finite distance between points are given in PEIERLS' article. For the present investigation some of these tables were recalculated with higher accuracy. The procedure is illustrated by the treatment of run no. 4 in table 1. On the basis of accuracy calculations PEIERLS recommends the number of periods over which the activity should be measured. This optimum measuring time depends on the ratio of background to exponential component, and with the background encountered in our case varies between 3 and 5 periods. This means that only the values of the first 8 to 14 channels should be used for the calculations. In run no. 4, where the initial value of the exponential distribution was 4.5 times the background, the optimum measuring time

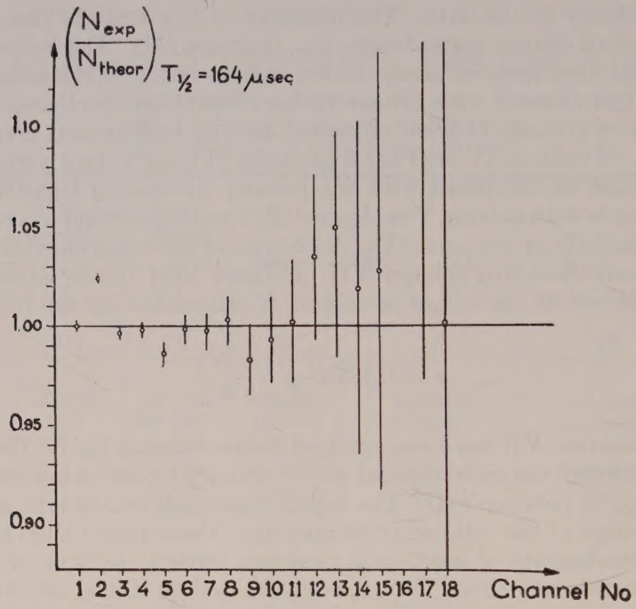

Fig. 5. Ratio of experimental values of run No. 4 series C to an exponential curve with a half-life of 164 μsec through the first point (logarithmic scale). Lengths of the vertical lines through the points indicate standard statistical errors of the experimental values.

corresponds to 10 channels. The remaining channels which were not used for the half-life determination were used chiefly for the determination of the background. Peierls' procedure was applied individually to each run of series C. For each value the standard error due to the statistical fluctuations of the number of counts was calculated from Peierls' tables as indicated in section D.

For series B the lower statistical accuracy of the runs did not warrant the same individual treatment as in series C. A rough check of the internal consistency of each run was made by plotting the number of counts after correcting them for the background on a semilogarithmic plot. The values for all the runs were then added together and the combined result was treated in the same manner as an individual run of series C.

The measurements of series A were of a more preliminary nature than those of the series B and C. Of the 10 channels completed only 7 yielded usable values because of various instrumental errors. For each run the values had to be corrected for the transient behaviour of the blocking oscillator which made the widths of the first channels up to 2 % larger than the following ones. The frequency of the blocking oscillator varied by about ±1 % and the data of each run were therefore corrected for the deviations of the channel width from the nominal value, 50 μsec. After these corrections had been applied to each run the results from all the runs were added and corrected for the background of chance coincidences, which was found by trial and error until the corrected values fitted a straight line on a semilogarithmic plot. From the slope of this line the half-life was found to be about 165 μsec. The standard error was estimated to be about 1.5 μsec from the scattering of the experimental points

Table 2

Run No. I	Beginning and end of run II	Length of run III	d μs IV	Half-life μs V	Stand. error μs VI	λ VII
A 1–6			50	165	3	
B 1–14, 17–19			77	164.40	0.28	
C 1	$\frac{10}{12}\ 23^{h}25^{m}$ — $\frac{11}{12}\ 12^{h}30^{m}$	$13^{h}05^{m}$	77	163.52	0.43	3.16
2	$\frac{11}{12}\ 13^{h}30^{m}$ — $\frac{12}{12}\ 15^{h}00^{m}$	$25^{h}30^{m}$	77	163.85	0.30	3.57
3	$\frac{12}{12}\ 15^{h}15^{m}$ — $\frac{12}{12}\ 17^{h}15^{m}$	$2^{h}00^{m}$	77	163.7	1.0	4.06
4	$\frac{12}{12}\ 19^{h}09^{m}$ — $\frac{13}{12}\ 11^{h}30^{m}$	$16^{h}21^{m}$	77	163.48	0.34	4.47
5	$\frac{16}{12}\ 01^{h}07^{m}$ — $\frac{16}{12}\ 16^{h}43^{m}$	$15^{h}36^{m}$	81	163.15	0.47	7.78
6	$\frac{16}{12}\ 17^{h}00^{m}$ — $\frac{17}{12}\ 11^{h}44^{m}$	$18^{h}44^{m}$	72	162.70	0.40	8.43
7	$\frac{17}{12}\ 12^{h}00^{m}$ — $\frac{18}{12}\ 16^{h}00^{m}$	$28^{h}00^{m}$	72	163.64	0.31	10.50
8	$\frac{18}{12}\ 16^{h}15^{m}$ — $\frac{19}{12}\ 15^{h}20^{m}$	$23^{h}05^{m}$	72	163.55	0.41	13.2
9	$\frac{20}{12}\ 16^{h}55^{m}$ — $\frac{21}{12}\ 09^{h}27^{m}$	$16^{h}32^{m}$	72	163.57	0.57	18.0
10	$\frac{21}{12}\ 09^{h}30^{m}$ — $\frac{22}{12}\ 09^{h}15^{m}$	$23^{h}45^{m}$	72	163.68	0.34	19.6
11	$\frac{22}{12}\ 09^{h}25^{m}$ — $\frac{23}{12}\ 10^{h}15^{m}$	$24^{h}50^{m}$	72	163.81	0.43	23.2
12	$\frac{23}{12}\ 10^{h}30^{m}$ — $\frac{27}{12}\ 15^{h}47^{m}$	$101^{h}17^{m}$	72	163.79	0.25	32.0
13	$\frac{27}{12}\ 16^{h}34^{m}$ — $\frac{28}{12}\ 10^{h}38^{m}$	$18^{h}04^{m}$	72	163.87	0.55	49.7
14	$\frac{28}{12}\ 12^{h}59^{m}$ — $\frac{29}{12}\ 10^{h}44^{m}$	$21^{h}45^{m}$	72			
		Weighted average:		163.69	0.10 (All measurements)	

Averages for different channel widths:

$d = 50$ μsec: 165 ± 3
$d = 72$ μsec: 163.60 ± 0.13
$d = 77$ μsec: 163.90 ± 0.16
$d = 81$ μsec: 163.15 ± 0.47

around this line. Systematic errors in the determination of the channel width and the uncertainty of the corrections are believed to increase this value to ±3 μsec.

The results of the half-life determinations from series A and B and from the 14 runs of series C are tabulated in Table 2, column V together with the calculated standard error (column VI), the channel width, d, (column IV) and the ratio λ of initial value of the exponential to the background (column VII). The variations of this latter factor with time during series C may be of some interest. The number of true coincidences is directly proportional to the source strength, and the number of chance coincidences is proportional to the square of the source strength (if the natural background of the counters can be neglected): thus the ratio should be inversely proportional to the source strength and should therefore increase exponentially with the half-life of the source.

Departures from this exponential behaviour are expected, when the source has decayed so much that the natural background of the counters becomes of importance. The data of column VII fit this predicted behaviour very well, which is considered as a further check on the proper functioning of the apparatus.

The weighted average of the half-life determinations in Table 2 was calculated, and the distribution of the individual values around this mean was studied and found to check roughly with what could be expected from the standard errors of the individual values. The value obtained for the half-life in this way is 163,69 μsec. The calculated standard error due to the statistical fluctuations was 0.10 μsec. In order to take into account the fact that an interpolated value was used for channel no. 2 this value should be increased to 0.13 μsec.

As there may be some doubt that the value of channel no. 1 is correct (see below, section G) in spite of the fact that the experimental points from this channel fitted very well with the theoretical curve through the rest of the points, the first two channels were omitted and the same calculations as before were repeated with the remainder of the experimental points. The weighted average of the values obtained by these new calculations is 163.80 ± 0.17 μsec, where the standard error as before is based only on the statistical fluctuations of the number of counts. The agreement with the first result is taken as a proof that channel 1 can safely be included in the calculation.

G. Sources of error

Errors orginating in the counters. The original exponential time distribution of the α-decay of RaC′ relative to the β-decay of RaC is modified by delays in the α- and β-counter. As is well known there is a spurious delay of the discharge in a G–M-counter which may amount to several microseconds. In the proportional counter itself there will be no corresponding effect but the finite rise-time of the amplifier for the pulses combined with the nonuniformity of the pulse heights will produce a spurious delay in the triggering of the discriminator of about 1 μsec. The effects of these spurious delays may be treated in connection with the finite resolution of the time analyzer, as their effect corresponds to a broadening of the triangular resolution function of the analyzer and a smoothing of the sharp corners.

After one of the β-counters has fired it will be insensitive during the deadtime of the counter which is of the order of 100 μsec. The β-pulse may be followed by spurious discharges during about 1 msec. These after-effects of the G–M-counters have no effect on the measurements because the time analyzer is not ready to accept another G–M-pulse until the switch has turned a complete revolution, i.e. after at least 1500 μsec. Even if some spurious discharges may occur after this time their only effect will be to increase slightly the background of random counts. If these after-effects were present in the α-counter, however, the measured value for the half-life would be affected. If the β-particles from the source decrease the sensitivity for α-particles for a certain deadtime after they have passed through the sensitive volume of the α-counter, the number of α-particles with small delays would be decreased and the measured life-time would appear too long. It is however not likely that the small ionization produced by the β-particles in the proportional region will affect the detection efficiency for a strongly ionizing α-particle. If there is a certain probability that an α-particle will produce spurious counts, with a distribution of delays which is roughly exponential, the period of the spurious counts will be superimposed on the original period and the apparent value would be too high or too low depending on whether the period of the spurious counts is larger or smaller than the period of the activity. No such spurious counts were detected by visual observation of the α-pulses on a scope using a triggered sweep, and they are probably non existent in proportional counters. They may introduce an error however if a G–M-counter is used for the detection of the α-rays, as pointed out by WARD (5).

The efficiency of the counting equipment was subject to large changes during the experiment, as no special care was taken to keep the operating voltage of the counters or the settings of the discriminators particularly constant, and as during such a long experiment with high counting rates aging effects may occur in the counters. Contrary to what would be the case if the time distribution were measured point by point this will not affect the value for the half-life, since all experimental points will be equally affected by a change in efficiency.

Resolution

The resolution functions of the different channels are illustrated by Fig. 3 C. These curves will be modified by the spurious delays in the counting equipment. Due to the effect of the prearranging circuit the resolution functions for the different channels will be identical with an accuracy which is more than sufficient for any physical measurement. The original time distribution, Fig. 3 E, is modified by the finite resolution of the system. It is easy to show however that the exponential behaviour of the distribution is conserved for all experimental points whose resolution triangles are completely within the region of the exponential decay. This is the case for all the channels except channels 0 and 1. The resolution triangle of channel 0 straddles the sharp rise of the time distribution curve at zero time. The number of counts registered in this channel will quite critically depend on the delays in the counting equipment and will be considerably below the exponential curve drawn through the remaining points. The tip of the left leg of the resolution triangle 1 will also extend into the region of non-exponential behaviour, if the delay between the event and the respective pulse to the time analyzer is larger for the α-particles than for the β-particles. The effect on the counts in the first channel is proportional to the square of the difference in delay, and is below 0.1 % if the difference is less than 3 μsec. Departure from the true exponential behaviour in channels 0 and 1 may further be caused by prompt coincidences. Prompt coincidences would occur if the pulses from the β-counter were electrically picked up by the α-pulse amplifier. This effect was eliminated by proper screening between the α- and β-counters. Prompt coincidences would also be caused by pick-up of external electrical interference in both the α- and β-counting channels. Any prompt coincidences would be distributed between channels 0 and 1 in the ratio of the channel width to the difference in delay between the α- and β-channels. The number of prompt coincidences was found to be too small to be observed by visual observation of the α-pulses on an oscilloscope, with the sweep triggered by the β-pulses. The fact that the experimental point obtained in the first channel fits very well with an exponential curve through the rest of the points may be taken as a further indication that the effect of prompt coincidences and of the delays on the counts in the first channel are negligible. The value obtained for the half-life when channel 1 is taken into account is also in close agreement with the value when only the channels after no. 2 are used for the calculation.

Resolving power of prearranging circuit

The prearranging circuit has a resolving time of the order of the channel width, and some counts may be lost in the circuit. At the counting rates used the loss will be negligible. It is further easily verified that the loss does not affect the half-life determination, because each β-pulse is followed by at most one correlated α-pulse.

Resolving power of the scalers and mechanical registers

The scale of 4 in each channel has a resolving power of about 25 μsec. As the prearranging circuit limits the number of pulses in each channel to one per revolution of the electronic switch, which occupies a time of the order of 1500 μsec, no pulses are lost in the scalers. The mechanical registers used in the channels are rather slow, and it was found experimentally that if random pulses were fed to the scale of 4 at a rate of 2000/min the mechanical registers will count 3 % more than the correct number. Below 1000 pulses/min the excess is less than 0.1 %. During the entire experiment the counting rate in any channel was always less than 500 p/min.

Erroneous functioning of the time analyser

In spite of the complexity of the electronic system and the long running time, the functioning of the apparatus proved satisfactory in general. Few defects occurred and were generally quickly localized and repaired during the periodical checks of the apparatus. The effect of these defects on the final results is probably negligible. A defect in a single channel was always quickly detected by the deviation of the corresponding point from the exponential curve through the other points and an interpolated value could be substituted for the erroneous one. Defective functioning of the gating circuit would change the form of the curve and was not found during the measurement. A change in frequency of the oscillator or frequency dividers would be the most serious error as it would give a systematic error in the half-life. The frequency of the crystal-controlled oscillator was compared at intervals with a standard frequency and the constancy was found to be of the order of 10^{-4}. The frequency ratio of the frequency dividers was checked about once a day and found to be unchanged. Any change in this ratio would be at least 1.2 %. As the result of no single run deviated by such a large amount from the mean value this error can hardly have occurred.

Statistical fluctuations of the number of counts in the channels

As the foregoing analysis of possible errors has failed to show any appreciable source of error, the accuracy of the result is believed to be limited entirely by the statistical fluctuations of the number of counts. This belief is supported by the fact that the scattering of the experimental points around the theoretical curves of fig. 5 is in good agreement with the expected statistical fluctuations. The effect of the normal statistical fluctuations on the half-life in the presence of a constant background has been treated by PEIERLS. An additional error might be expected in the determination of the background which was not measured directly but extrapolated from the experimental points in the manner described. However the points used for the background determination were chiefly the last 5 or 6 points, while, following PEIERLS' recommendation only the first 8–14 points were used for the determination of the half-life. The two determinations are therefore almost independent of each other and the effect of the interrelation of the two determinations on the statistical error of the half-life can be neglected.

The scattering of the half-life values obtained in different runs, using 4 different channels widths 50, 72, 77 and 81 μsec, and counting rates and activity to background ratios which vary by a factor of 20 agrees well with the fluctuations which are to be expected according to PEIERLS statistical treatment. No systematic errors have been found in the apparatus and methods used. It is believed that any unknown systematic error which might be present would affect differently runs made under widely different conditions and thus cause a systematic spread of the results. It is therefore felt that unknown systematic or accidental errors could at most be of the same order of magnitude as the error due to the statistical fluctuations. In order to take account of these possible errors it is considered that the standard error in the half-life determination should be increased to ± 0.2 μsec. from the value of ± 0.13 μsec, which was due to statistical fluctuations of the number of counts alone.

H. Discussion

The obtained value for the half-life of RaC′, 163.7 ± 0.2 μsec, is not inconsistent with the value obtained by JACOBSEN and SIGURGEIRSSON, 155 ± 5 μsec, using the delayed coincidence method. However the results obtained by the coincidence method of variable resolution differ markedly from the present value. A number of measurements with this method have yielded results in good agreement with each other at about 145 μsec, the accuracy of most of the measurements being about 5 μsec. The discrepancy with the present value seems too large to be purely accidental and one is led to believe that there may be systematic errors inherent in the coincidence method of variable resolution. It is suggested that this point should be further investigated, as the errors may affect all half-lives determined by this method.

The use of filters for the α- and β-radiations would influence the experimental values if several energy groups are present. Long range α-particle groups are known to exist in the α-spectrum of RaC′. They are emitted from excited states of RaC′. In principle the result of the present and previous measurements would lead to a time distribution which is a superposition of exponentials with the half-lives of the ground state and excited states. The excited states of RaC′ are believed to have extremely short half-life for radiative decay to the ground state. With equipment available at present the exponential decay of the excited states can not be resolved, with the low resolution used, and the long range α-particles would only contribute to the prompt coincidences but would not influence the half-life determination. Their number is furthermore small compared to the normal α-rays (about 10^{-4}). The present measurement as well as the previous ones should therefore yield the half-life of the ground state of RaC′, independent of the amount of absorbers used in the path of the α- and β-rays.

The method used in the present investigation, the multichannel differential method, seems to be capable of higher accuracy than previous methods, especially after the modification described as the pre-arranging circuit. As the limiting factor is still the statistical fluctuations of the number of counts, the accuracy of the present value can be somewhat increased by more prolonged measurements. The fact that a number of internally consistent values are obtained in a single run and that the results are unaffected by changes in counting rates during the measurement makes the method particularly suitable for measuring delayed coincidences on substances which decay rapidly. The multichannel feature further decreases the time needed for a given experiment. This is particularly useful in a search for rather long half-lives with the delayed coincidence method. It is estimated that half-lives of the order of 1 sec. could still be measured.

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